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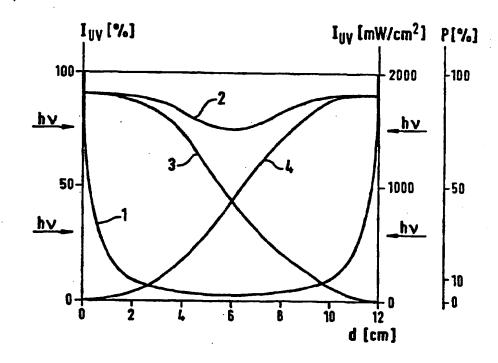
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(54) Title: METHOD AND DEVICE FOR POLYMERISING BINDERS IN FIBROUS MATERIALS ESPECIALLY MINERAL WOOL INSULATION

#### (57) Abstract

The invention concerns a method and a device for polymerising substances in fibrous materials, in particular binding agents in mineral wool materials for insulation purposes, wherein the fibrous material having a given thickness (d) is subjected to UV irradiation in order to polymerise the substance having the form of a prepolymer impregnating the fibres, said irradiation of the material is carried out at a sufficiently high intensity level (IUV) to bring about, at a maximum depth (d) of intended polymerisation, a residual intensity exceeding such threshold value at which polymerisation of the selected binding agent under influence of UV radiation is ensured within a given time limit, such time limit, however, precluding any undesirable degradation, due to the effects of the radiation, of organic substances in the portion at the



surface of said material, and wherein the duration of irradiating a given surface unit of said material is kept within said time limit.

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#### **Description**

METHOD AND DEVICE FOR POLYMERISING BINDERS IN FIBROUS MATERIALS ESPECIALLY MINERAL WOOL INSULATION

The invention concerns a method for polymerising substances in fibrous materials, in particular binding
agents in mineral wool materials for insulation purposes,
wherein the fibrous material which has a given thickness
and which, in a given case, may be continuously moving in
a line of production, is subjected to UV irradiation in
order to polymerise the substance having the form of a
prepolymer impregnating the fibres, and a device for
polymerising substances in fibrous materials, such as
particularly a binding agent in a mineral wool material
for insulation purposes, comprising at least one UV
radiation source.

Such a technique is known from US-A-5 275 874. It is the purpose of the known teaching to uniformly polymerise the material across its thickness. In view of the fact 25 that the energy released inside the material by the UV rays exponentially declines with increasing distance from the material surface, a considerable treatment duration is required in order to achieve maximum uniformity of exemplary case curing. In the of US-A-5 275 874, radiation intensity across the thickness of the material 30 drops to 1% of the incident radiation as the material passes under a plurality of consecutively arranged UV lamps, essentially in analogy with a passage through the curing furnace in conventional thermal curing.

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With such a treatment, experience within the framework of the present invention teaches that the

surface-side organic constituents of the mineral wool material are inevitably scorched or carbonised. In view of the rapid drop in intensity, i.e. the power released per surface unit, with the depth below the irradiated surface, the required energy input is so high as to bring about carbonising or scorching of the surface-side organic constituents, e.g. the binding agent, at the irradiated surface of the mineral wool material.

10 Correspondingly, the invention is based on the object of providing a method according to the preamble of claim 1 whereby the desired polymerisation is made possible even in considerable material depths below the irradiated surface without the occurrence of any undesirable decomposition effects at the material surface.

This object is achieved in that irradiation carried out at a sufficiently high intensity level to bring about, at a maximum depth intended 20 polymerisation, residual intensity exceeding а threshold value at which polymerisation of the selected binding agent under the influence of UV radiation is ensured within a given time limit, such time limit, however, precluding any undesirable degradation due to the effects of the radiation of organic substances in the 25 portion at the surface of the material, and in that the duration of irradiating a surface unit of the material is kept within this time limit. In the case of a fibrous material continously moving in a production line, this duration will also be selected so as to be compatible 30 with the velocity of the production line.

In the course of research of the work relating to the present invention it was found that the materials eligible as a prepolymer exhibit threshold values for the intensity level, i.e. the irradiated power per area unit,

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above which polymerisation takes place within a very short time period, virtually instantaneously. Increasing the intensity level above this threshold value does not appreciably accelerate polymerisation, without, however, unfavorably affecting it. The temperature increase at the irradiated surface leading to the occurrence of degradation effects, on the other hand, is essentially proportional to the duration of irradiation, with the velocity of the temperature increase naturally increasing as a function of the levels of intensity.

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By the application of UV radiation, heat is released as a function of the radiation intensity, either due to the heat radiation concurrently emitted by the UV source, or due to energy losses of the UV radiation which is thus transformed into thermal radiation; these effects appear most clearly at the surface of the material, with the highest temperatures occurring there and possibly bringing about decomposition effects once a given temperature limit is exceeded.

These effects which occur in parallel, however with a different course over time, are used in the present invention firstly, the threshold level in that intensity is determined at which polymerisation of the material occurs rapidly, e.g. within 0.2 s. The area unit in question is then irradiated for this duration, and the surface intensity level of irradiation may be raised to such an extent that the rise of temperature during this predetermined time span of e.g. 0.2 s still remains below a given temperature limit above which degradation effects have to be expected. This maximum intensity, which may be applied within the predetermined short period of time, due to the intensity drop inside the material, in turn limits the depth within which the intensity remains above

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the threshold value and within which a virtually homogeneous polymerisation is therefore possible.

As each area unit is consequently exposed to a high intensity level radiation for a short time period only, irradiation may be carried out in a limited space to save production line space. In the case of a continuously moving web of material in the production line, the time span of irradiation of each area unit has to be selected so as to be compatible with the velocity of the line, generally between 0.1 and 1 m/s.

If the intensity level of irradiation is in excess of 500 mW/cm², in particular in excess of 1 W/cm², and in a particularly preferred manner exceeds 2 W/cm², then commercially available UV sources may be applied for cost efficiency on the one hand while nevertheless enabling high surface intensity levels on the other hand. As is known per se, UV sources for radiation in the wavelength area above 250 nm, preferably above 310 nm are preferred in view of the selectively elevated transmissivity of the mineral wool material in this area.

The duration of irradiating a surface unit is prefer-25 ably shorter than ten seconds, preferably shorter than 1 s, in particular shorter than 0.5 s.

In many conventional production methods, the mineral fibres are produced from molten glass, provided with the substance, e.g. the prepolymer of the binding agent, in the course of deposition as a layer of wool, and then the substance, e.g. the binding agent, is cured. In such a case, the mineral wool material still has a relatively elevated temperature when the substance is being cured, such that the temperature rise on the surface side to the temperature limit under the influence of the irradiation

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starts out from a relatively high initial temperature. Particularly in such a case it is preferred to cool the mineral wool material to a temperature as low as possible before irradiation to thus allow for a larger permissible increase of temperature due to the irradiation treatment. Cooling measures concurrent with irradiation, however, in view of the short time period of irradiation do not have an effect that would justify the expenditure associated therewith. Applying the method of the invention, on the other hand, is particularly advantageous with all those production methods where the mineral wool material is present at a lower temperature, preferably at ambient temperature, before curing. This is the case e.g. when the binding agent is introduced in the form of vapor or of an aerosol, as is known from DE-A-44 06 863 and DE-A-44 10 020, respectively.

The proportion of oxygen contained in the fibrous material, which in usual processing is at about 21% in the surrounding atmosphere, is preferably reduced during irradiation to less than 10%, better even 5%, and in a particularly preferred manner to less than 1%. In usual polymerisation making use of radicals, this measure prevents the free radicals of the substance to be polymerised, which are generated by the radiation, from being occupied by oxygen and thus prevents a failure of polymerisation in those places. This is particularly important with very thin film thicknesses of monomers and low intensity of irradiation.

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Irradiation is preferably applied to both major surfaces of the material, such that the radiation energy penetrates across the thickness of the material from both sides. The penetrating depth of the radiation may thus be more than doubled when both penetrating depths overlap in the centre of the material. This overlapping effect is

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made particularly good use of if both sides are irradiated concurrently, such that in a certain location inside the material the energies from irradiating both sides are present simultaneously.

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Polymerisation or curing does not necessarily have to be homogeneous across the thickness of the material. Especially in the case of bilateral irradiation, it may be perfectly sufficient if the layers of material adjacent to the surface are cured and a more or less large inner width of considerably reduced polymerisation remains. In the case of curing a binding agent, the desired increase in mechanical strength is thus effected in the outer portions, such that resistance against bending, which frequently is of primary importance, hardly suffers from a lower mechanical strength in the central portion of the panel. Incomplete curing frequently also acceptable where other properties than the bending resistance, e.g. surface tightness, surface strength, or the like are of importance. When other substances than binding agents are polymerised, homogeneous polymerisation across the thickness may prove to be favourable depending on the function of the substance and the use of the product.

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In the case of non-homogeneous polymerisation across the thickness it may also be favourable, in accordance with the teachings of DE-A-44 06 863 or DE-A-44 10 020, to already introduce the substance into the material in a non-homogeneous distribution. E.g. in the case of binding agent, a higher binding agent concentration may thereby be achieved in the portions near the surfaces than in the central portion of the panel, resulting in a production with a desired optimum use of a certain amount of binding agent.

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For the prepolymer, in particular multi-functional acrylic and metacrylic compositions are suited, as is known per se. Further usable substances are prepolymers form of monomers, oligomers, comprising polymerisable, unsaturated functional groups such as acrylate, methacrylate, vinyl, vinyl ether, allylic, or maleate groups which are capable of reacting in the sense of a chain extension and/or cross-linking. binding agent may be a mixture from compositions and contains a photo-initiator in order to enable polymerisation through UV light. Prepolymer mixtures comprising epoxy acrylate were found to be particularly preferred with the present invention.

- A device according to the invention serving to implement the method of the invention is characterised in that means for focussing the UV radiation to at least one focussed beam of small width are provided.
- Due to such focussing, a desired high level of surface intensity may be achieved.

Particularly in polymersation at a running production line it may be preferred if focussing is effected in a linear shape, e.g. extending across the width of the production web. The width of the linear focussed beam may then be coordinated with the transport velocity of the production web to result in a desired duration of irradiating each surface unit without the presence of any moved parts.

As an alternative, however, focussing may also be effected in the shape of a point, with the thin high-intensity beam thus formed sweeping the material in a desired predetermined pattern. The substance may thus be cured in fibrous material having an uneven surface in the case of

8

shaped articles, e.g. pipe sections having the shape of a hollow cylinder. This movement of the focussed beam may, for example, even be obtained at a quite limited expenditure by means of a sweeper mirror. The high intensity or power density of such a beam focussed in the shape of a point enables large penetrating depths of polymerisation for substances having particularly short polymerising times. Particularly, such sweeping movement with a thin allows adaptation to different a good velocities. For certain cases of use, it may suffice to sweep only a fraction of the total surface area of the fibrous material with the beam and to allow for gaps in between, within which polymerisation is not necessary or does in fact occur as a distant effect of the adjacent irradiation.

In a preferred manner, the focussed beam is directed at a finite focal point, i.e. tapering into a point instead of consisting of parallel rays. Thanks to suitable focussing, the focal point may be positioned at a desired depth of the material so that a relatively higher intensity on a smaller surface is obtained there, even though attenuated by scattering at fibres located further upwards. Such a formation of the focussed beam is particularly suitable in the case of a line-shaped focussed beam.

Further details, features and advantages of the invention can be seen from the following description of embodiments by making reference to the drawing, wherein:

Fig. 1 shows a typical function of the intensity level versus the wall thickness of a mineral wool product subjected to bilateral UV irradiation,

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- Fig. 2 shows the rate of conversion by polymerisation versus time at various intensity levels,
- Fig. 3 shows the surface heating of the mineral 5 wool material at various intensity levels,
  - Fig. 4 shows the relation of intensity level and polymerisation across the wall thickness of a mineral wool product treated according to the invention with bilateral UV irradiation,
    - Fig. 5 shows a schematically simplified representation of a device according to the invention having linear focussing of the UV radiation, and

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Fig. 6 shows a schematically simplified representation of a device according to the invention having a point-shaped focussed beam on the mineral wool material.

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In order to facilitate comprehension, the same reference signs are used for identical or corresponding parts or graphically represented functions, respectively.

- 25 Fig. 1 illustrates a typical function of the local intensity level of UV radiation versus the thickness of a fibrous material, for example a mineral wool product subjected to bilateral UV irradiation of its surface.
- As the abscissa or X axis, the wall thickness or depth d of the mineral wool product of, in the case of the example, a wall thickness of 12 cm is shown. The left ordinate or Y axis shows the intensity level of the UV radiation  $I_{\rm UV}$  of 0 to 100% and the right ordinate or Y axis shows the intensity level of the UV radiation  $I_{\rm UV}$  of 0 to 2000 mW/cm<sup>2</sup>. In the example shown, 2000 mW/cm<sup>2</sup>

correspond to the 100% intensity level of the UV radiation.

On the right side of the right ordinate, on the one hand, and on the left side of the left ordinate, on the other hand, arrows are drawn which, pointing towards the middle of the Figure, symbolize the incident UV radiation having the energy hv. Since irradiation occurs from both sides of the mineral wool product, the intensity level 10 over the thickness d illustrated by curve approximately symmetrical to the centre line so that initially only the left half of the diagramm shall be considered herebelow. It can be seen that the intensity level I<sub>UV</sub> sharply drops already after a relatively short 15 penetration depth d. From an initial intensity level Im. of 100% at the mineral wool surface, the intensity level  $I_{UV}$  drops, in this example, to below 50% already after less than 1 cm penetration depth, after a penetration depth of approximately 2 cm already to below 10% and, in 20 the middle portion of the mineral wool product, amounts to only below 5% of the intensity level  $\mathbf{I}_{\mathbf{UV}}$  incident on the surface. This sharp decline of the intensity level  $I_{UV}$  over the penetration depth is, on the one hand, caused by the low transmission of the UV radiation 25 through the mineral wool material and, on the other hand, a result of a conversion of UV radiation energy to thermal energy already in the uppermost layers of the mineral wool material, without a related polymerisation.

A first evaluation of curve 1 of intensity as shown leads to the result that a satisfactory polymerisation of substances in fibrous materials must be restricted to portions of material near the surfaces since the UV radiation intensity level  $I_{UV}$  decreases too rapidly, thus failing to provide a sufficiently high residual intensity in the centre portion for accomplishing satisfactory

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polymerisation there. If, therefore, the intensity level  $I_{UV}$  of the radiation were increased to such an extent that the centre portion of the fibrous material can still be successfully polymerised, this would entail such high intensity levels at the surface that damage to the surface-side material, for example carbonisation or scorching of the surface-side organic constituents, e.g. the polymerisable substance or a binding agent, could not be avoided.

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In Fig. 2 the function of the polymerisation of substances in fibrous materials versus the time at various intensity levels of the UV radiation is shown. At the abscissa or X axis, the irradiation time t in seconds is shown from 0 to 1.5 s. The left ordinate or Y axis shows the related conversion rate by polymerisation P from 0 to 100%. The curves  $I_{\rm UV1}$  to  $I_{\rm UV7}$  show various intensity levels  $I_{\rm UV}$  of the UV radiation of, for example,  $I_{\rm UV1}$  = 2 mW/cm²,  $I_{\rm UV2}$  = 5 mW/cm²,  $I_{\rm UV3}$  = 11 mW/cm²,  $I_{\rm UV4}$  = 25 mW/cm²,  $I_{\rm UV5}$  = 55 mW/cm²,  $I_{\rm UV6}$  = 128 mW/cm², and  $I_{\rm UV7}$  = 220 mW/cm². Thus, the intensity level  $I_{\rm UV}$  increases from  $I_{\rm UV1}$  to  $I_{\rm UV7}$ .

All seven curves exhibit typical common features. Those are, firstly, a common point of origin in the 25 origin 0 of the graph. After a short delay, this common point of origin is followed by an ascent of the curve with a positive gradient dP/dt, which is followed by an almost linear area of ascent with a constant positive 30 gradient dP/dt. This area of linear ascent, furthermore, is followed in all shown curves by a decreasing ascent, i.e. having a decreasing positive gradient dP/dt, to arrive at an almost horizontal linear section without further ascent, i.e. having a constant gradient dP/dt of approximately 0. The limit of the conversion rate by 35 polymerisation P reached in this constant section can be

designated as a saturation limit of the polymerisation P occurring at a specific intensity level  $I_{\sigma V}$  of radiation.

The steep gradient of the linear ascents in the front 5 portion of the curves increases with increasing intensity  $\mathbf{I}_{\mathbf{UV}}$  of the UV radiation, and is not very distinct in the case of the curve for  $I_{\mathrm{UV1}}$  with 2 mW/cm², whereas a very steep gradient is present for the curve I<sub>UV7</sub> with 220 mW/cm<sup>2</sup>. The arc of transition from the steep ascent 10 in the front portion of the curves to the horizontal, almost linear portion of the curves is not very distinct in the case of the two intensity levels  $I_{UV1}$  and  $I_{UV2}$ , in the case of the intensity level  $I_{UV3}$ it is discernable, and the intensity levels  $I_{UV4}$  to  $I_{UV7}$  exhibit a clearly distinct arc of transition from the sharp 15 ascents to the constant portions of the curves. Moreover this arc of transition occurs at an earlier point in time with increasing intensity level Inv.

This means that, in the cases of lower intensity 20 levels  $I_{UV}$  of UV radiation, the mineral wool material has to be irradiated for a relatively longer time until no further increase of polymerisation, which is low at any rate, occurs. However, if a relatively high UV radiation is used for irradiating the mineral wool material, a high 25 degree of polymerisation is achieved already within a very short period of time which very quickly reaches a maximum and can then be increased no more but develops This means that there is limit irradiation time for each significant intensity level of 30 radiation, within which a maximum polymerisation reached and no further increase of polymerisation can be achieved by exceeding said limit through irradiation.

In the example shown, a sharp increase of polymerisation occurs at an intensity level  $I_{\text{DV7}}$  of 220 mW/cm² already after approximately 0.05 s and reaches its maximum value already after a duration of irradiation of 0.2 s - 0.3 s, in this example of approx. 80%. This shows that, at a sufficient intensity level of irradiation, a high-level maximum of polymerisation can be reached already after a short time of irradiation.

10 The surface heating of the fibrous material or of the mineral wool material at various intensity levels of the UV radiation is explained thereafter in more detail with reference to Fig. 3. The abscissa or X axis represents time t in seconds. The ordinate or Y axis shows the temperature of the surface of the mineral wool material in °C. The horizontal dashed line at 20°C symbolises the ambient temperature as a reference value. In the present example starts from the assumption that the mineral wool material is provided at a temperature of approx. 20°C before irradiation. The second horizontal dashed line at 200°C shows the upper limit of the thermal load on the organic constituents, such as the polymerisable substance or the binding agent of a usual mineral wool material at its surface. In the graph itself, the function of 25 temperature versus time is shown for two different intensity levels  $I_{\overline{UV1}}$  and  $I_{\overline{UV2}}$ , of which  $I_{\overline{UV1}}$  is the smallest one in the case of the example 11  $mW/cm^2$ , and  $\mathbf{I}_{\mathbf{UV2}}$  the highest intensity level in the case of the example 2000  $mW/cm^2$ . It can be seen that the temperature of the surface of the mineral wool material increases 30 almost proportionally with time at a constant intensity level of the UV radiation. This linear temperature increase depends on the intensity level  $\mathbf{I}_{\mathbf{UV}}$  in a way that the gradient of this straight temperature line increases 35 with increasing intensity levels. If the mineral wool material is irradiated with high intensity levels of UV

radiation, the temperature limit of e.g. 200°C, at which damage occurs at the surface of the mineral wool material, is already reached after a relatively short time.

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The new knowledge about these interrelationships can be put to use within the teaching of the present invention by reducing the irradiation periods when irradiating mineral wool materials to such an extent that the arc of transition from the steep ascent to the maximum of polymerisation may just be reached within this period. This allows the use of intensity levels which, although they would lead to damage at the surface after longer irradiation, do not cause damage at the surface at such short irradiation times and still allow the introduction of a sufficient intensity into the centre of the material in order to also allow a satisfactory polymerisation P within such irradiation period.

Together with the physical relations shown in Figs. 1 and 2, it becomes clear that there is a working range depending on the intensity level I<sub>UV</sub> of UV radiation, the thickness d of the mineral wool product, the irradiation period t and the shown temperature limit, wherein a maximum and rapid polymerisation of substances in fibrous materials is possible without having to expect surface damage.

The mineral wool material may be present at a high 30 temperature prior to irradiation due to preceding treatment steps. Further heating due to the irradiation may then cause the temperature limit to be exceeded relatively rapidly and thus cause thermal load of the organic constituents such as of the polymerisable 35 substance or of the binding agent of a usual mineral wool material at the surface thereof. It may thus be envisaged

to cool the mineral wool material prior to irradiation e.g. by means of suitable devices as they are proposed e.g. in DE-A-44 06 863 or DE-A-44 10 020, respectively, whereby a rapid temperature rise due to irradiation to values beyond the temperature limit and consequently damage to the surface may be precluded.

The exemplary relationship according to the present invention between variations of intensity level on the one hand and polymerisation on the other hand, and the 10 thickness d of a mineral wool product at bilateral UV irradiation is shown in Fig. 4. The abscissa shows in the same way as in Fig. 1 the thickness d of a mineral wool product of, in the case of the example, 12 cm. At the 15 left ordinate, the intensity level  $I_{UV}$  from 0 to 100% and at the right ordinate the intensity level  $I_{\mbox{\scriptsize UV}}$  from 0 to 2000 mW/cm2 is shown. In this, the intensity level of 100% corresponds to the 2000 mW/cm2. Additionally the polymerisation P from 0 to 100% is shown at the right 20 ordinate. Like in Fig. 1, the arrows hv from the right from the left symbolize the energy irradiation with UV radiation. Curve 1 again shows the variation of the intensity level over the thickness d corresponding to Fig. 1. A curve 3 shows the variation of 25 polymerisation P at irradiation from the left and a curve 4 shows the variation of polymerisation P at irradiation from the right. If irradiation takes place from both sides, the development of polymerisation P results from a superposition of curves 3 and 4, which is graphically 30 represented by curve 2. In the exemplary case as shown, curves 3 and 4 are approximately mutual mirror images with the center of the mineral wool material as their such that the curve 2 is also approximately symmetrical with the center portion.

following, curves 1 und 3 are considered, on the basis of which the relation between the variation of the intensity level and polymerisation P shall be shown. As long as the intensity level  $I_{TV}$ , which sharply drops already after little penetration depth in curve 1, still exceeds a sufficiently high limit, an almost maximum polymerisation P takes place. If this limit is not reached, curve 3 drops as well and finally approaches zero. The same holds true for curve 4 at 10 irradiation from the right. If the explanations given in respect of Figs. 1 to 3 are taken into consideration, it becomes clear that an almost uniformly polymerisation can be achieved in the mineral wool material over its complete thickness at a sufficiently high 15 starting intensity of the UV radiation at the surface of the mineral wool product, which then, typically, rapidly decreases, however not further than to a specific intensity limit at which polymerisation still noticeably occurs, and at a correspondingly chosen duration of 20 irradiation. In this manner it would be possible to polymerise mineral wool materials across their entire thicknesses. However, it is also possible, for example by acting on the exposure time, to cause substances in these mineral wool materials to polymerise only in the vicinity 25 of their surfaces and to cause no polymerisation or only a reduced polymerisation in their center portions.

Any unpolymerised parts of the substance within the fibrous material are exposed to ageing due their reacting with atmospheric oxygen and other reactants, whereby their reactivity is eliminated and the substance is made inert.

Fig. 5 shows a schematically simplified 35 representation of an exemplary embodiment of a device according to the invention having linear focussing of the

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UV radiation. A UV radiation source 10, preferably a linear UV radiation source, is located in the first focal point of a reflector 12 whose cross-section preferably has the form of a parabola or an ellipse. radiation emitted by the UV radiation source 10 focussed onto its second focal point 14 by means of the reflector 12. The second focal point 14 is located in the vicinity of the surface of the mineral wool material 16. to be irradiated or within the mineral wool material. The mineral wool material 16 may be provided as mineral wool material web and is, for example, transported on a conveyor device, not shown, underneath at least one UV radiation source 10 and/or above at least UV radiation source 10, and thus swept by the linearly focussed beam 18 of a minimum width b approximately at right angles to the direction of transport.

Fig. shows a schematically simplified representation of a device according to the invention 20 producing a point-shaped focussed beam on the mineral wool material. The UV radiation source 10 is, in this case, point-shaped and arranged in the first focal point of an approximately semi-ellipsoidal reflector 12. means of this semi-ellipsoidal reflector 12, UV radiation of the radiation source 10 is focussed in the second 25 focal point 14. The second focal point 14, on the other hand, is located in the vicinity of the surface or within the mineral wool material 16, which may, for example, be formed as a three-dimensional shaped article, and whose surface contour is swept by the focussed UV beam 18 under 30 an angle of penetration of approximately 90° to the planar normal of the respectively irradiated planar element. This can be accomplished by means of a suitable moving equipment for the UV radiation source 10, not shown, and/or by means of movable mirror units, also not 35 shown.

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The devices of Fig. 5 and 6 use finite focal points at the level of the mineral wool material to be treated. However, quite similar effects may also be obtained when using thin beams with infinite focal points, i.e. parallel rays.

The oxygen content in the fibrous material during the treatment is to be reduced preferably to below 10% or even more preferably to below 5% or even most preferably to below 1%, whereby the case in which the free radicals of the substance to be polymerised, which are produced by the UV radiation by means of the photo-initiators, are reacted with oxygen and a polymerisation at this position is prevented may be avoided practically altogether.

In case of sufficient reduction of the atmospheric oxygen content in the fibrous material, a more complete polymerisation of the substance can thus be achieved. It is a further advantage of the use of an inert atmosphere to reduce the possible formation of ozone since in a mostly inert atmosphere, oxygen molecules which might be partly split by the energy of the UV radiation to form oxygen radicals, and which might then form ozone are no longer present.

The reduction of the oxygen content may be obtained in a simple manner by flushing the mineral wool material with another gas, for example nitrogen, or in a more economic manner with carbon dioxide or water vapour.

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#### Claims

1. Method for polymerising substances in fibrous 5 materials, in particular binding agents in mineral wool materials for insulation purposes, wherein the fibrous material which has a given thickness (d) and which, in a given case, may be continuously moving line of production, is subjected 10 irradiation in order to polymerise the substance having the form of a prepolymer impregnating the fibres,

#### characterised in that

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irradiation of the material is carried out at a sufficiently high intensity level ( $I_{trv}$ ) to bring at maximum depth of intended polymerisation, a residual intensity exceeding such threshold value at which polymerisation of the selected binding agent under the influence of UV radiation is ensured within a given time limit, such however, precluding any undesirable degradation, due to the effects of the radiation, of organic substances in the portion at the surface of said material, and in the case of fibrous material continuously moving in a line of production being compatible with the velocity of the line; and

- in that the duration (t) of irradiating a given surface unit of said material is kept within said time limit.
- 2. Method according to claim 1, characterised in that said intensity level  $(I_{UV})$  at the surface of said

mineral wool material exceeds 500 mW/cm<sup>2</sup>, preferably exceeds 1 W/cm<sup>2</sup>, and in particular exceeds 2 W/cm<sup>2</sup>.

- 3. Method according to claim 1 or 2, characterised in that the wavelength of the UV radiation utilised is in excess of 250 nm, preferably of 310 nm.
- 4. Method according any one of claims 3, characterised in that the duration (t) of 10 irradiating said surface unit is shorter than ten seconds, preferably shorter than 1 s, and in particular shorter than 0.5 s.
- 5. Method according to any one of claims 1 to 4,

  15 characterised in that the surface of said mineral wool material is cooled prior to irradiation.
- 6. Method according to any one of claims 1 to 5, characterised in that the proportion of oxygen contained in the atmosphere inside said mineral wool material is reduced to less than 10 %, preferably to less than 5 %, in particular to less than 1 %.
- 7. Method according to any one of claims 1 to 6,

  25 characterised in that irradiation is applied to both major surfaces of said material.
  - 8. Method according to claim 7, characterised in that both sides are irradiated concurrently.
  - 9. Method according to claim 7 or 8, characterised in that a low-polymerisation zone is provided in the central region of said material.
- 35 10. Method according to any one of claims 1 to 9, characterised in that a mixture of substances

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comprising an epoxy acrylate is used as the prepolymer for forming a binding agent.

- 11. Device for polymerising substances in fibrous materials, such as particularly a binding agent in a mineral wool material for insulation purposes, comprising at least one UV radiation source (10), characterised in that means (12) are provided to focus the UV radiation into at least one focussed beam (18) of small width (b).
  - 12. Device according to claim 11, characterised in that said focussed beam (18) is shaped to be linear, and is preferably kept stationary.
- 13. Device according to claim 11, characterised in that said focussed beam (18) has the shape of a point rapidly sweeping said mineral wool material (16) from one side to the other so as to irradiate preferably all of its surface area.
  - 14. Device according to any one of claims 11 to 13, characterised that said focussed beam (18) has a focal point (14) located within said mineral wool material (16) to be treated.
- 15. Device according to any one of claims 11 to 14, characterised in that UV radiation sources (10) are positioned on both sides of the major surfaces of said mineral wool material (16).





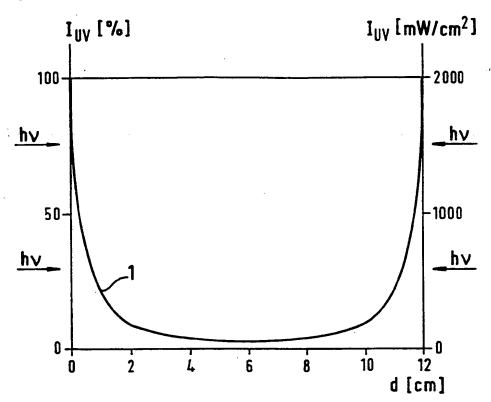
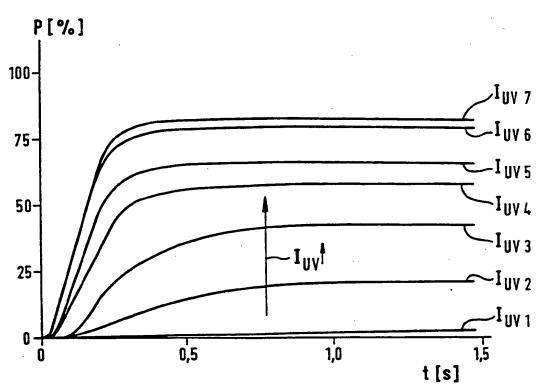


Fig. 2



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Fig. 3



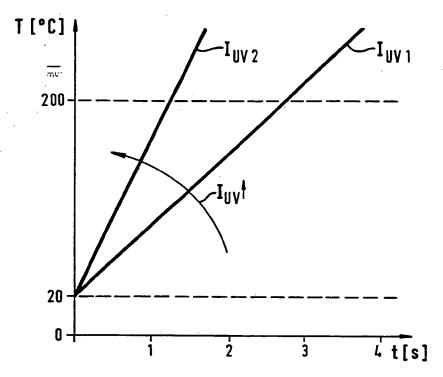
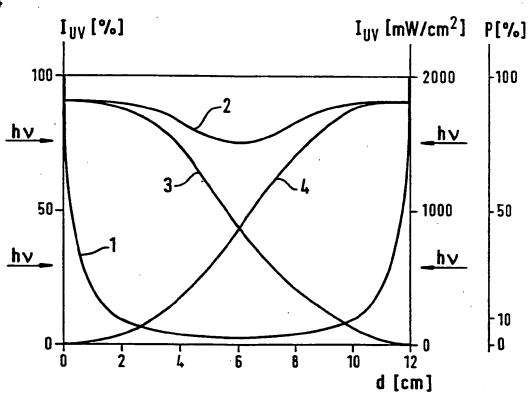
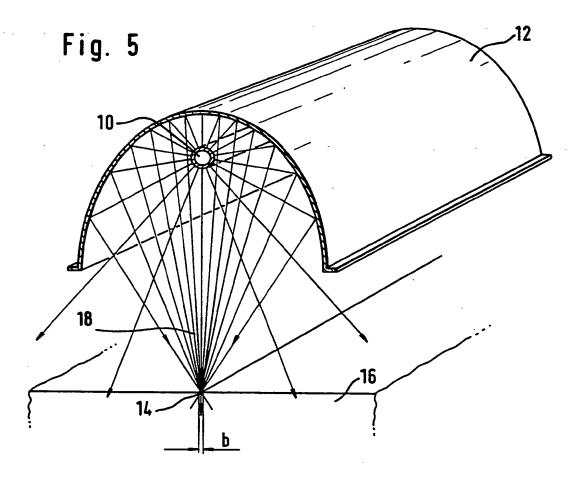
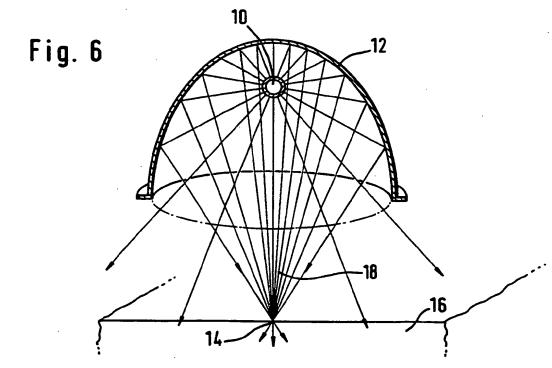


Fig. 4







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#### INTERNATIONAL SEARCH REPORT

Interne al Application No

PCT/EP 95/02325 A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 D04H1/64 D06M1/ D06M10/00 According to International Patent Classification (IPC) or to both national classification and IPC Minimum documentation searched (classification system followed by classification symbols) DO4H DO6M IPC 6 Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched Electronic data base consulted during the international search (name of data base and, where practical, search terms used) C. DOCUMENTS CONSIDERED TO BE RELEVANT Citation of document, with indication, where appropriate, of the relevant passages Relevant to claim No. US,A,5 275 874 (K BRANDENBURG ET AL) 4 1,3 Y January 1994 cited in the application see the whole document Y MELLIAND TEXTILBERICHTE 1,3 vol. 74, no. 5, May 1993 HEIDELBERG DE, pages 428-434, 'PIGMENTIERTE UV-HÄRTENDE M MARSMAN ET AL BESCHICHTUNGEN FÜR TEXTILANWENDUNGEN' see page 432 X DE,A,29 37 081 (UNISEARCH LTD) 20 March 1,11 1980 see claims; tables 14,17 Further documents are listed in the continuation of box C. Patent family members are listed in annex. Special categories of cited documents: T later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the 'A' document defining the general state of the art which is not considered to be of particular relevance invention earlier document but published on or after the international "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) involve an inventive step when the document is taken alone "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such docu-"O" document referring to an oral disclosure, use, exhibition or ments, such combination being obvious to a person skilled in the art. document published prior to the international filing date but later than the priority date claimed "&" document member of the same patent family Date of the actual completion of the international search Date of mailing of the international search report **20.** 10. 95 29 September 1995 Name and mailing address of the ISA Authorized officer European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax (+31-70) 340-3016

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# INTERNATIONAL SEARCH REPORT

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PCT/EP 95/02325

L	tion) DOCUMENTS CONSIDERED TO BE RELEVANT		
Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.	
X	DE,A,21 24 691 (IMPERIAL CHEMICAL INDUSTRIES) 9 December 1971 see page 5; claims	11	

Form PCT/ISA/210 (continuation of second sheet) (July 1997

## INTERNATIONAL SEARCH REPORT

...ormation on patent family members

Intern al Application No
PCT/EP 95/02325

Patent document cited in search report	Publication date	Patent family member(s)		Publication date
US-A-5275874	04-01-94	NONE		
DE-A-2937081	20-03-80	AU-B- AU-B- GB-A,B US-A-	536517 5071079 2036039 4401688	10-05-84 20-03-80 25-06-80 30-08-83
DE-A-2124691	09-12-71	GB-A-	1349058	27-03-74

Form PCT/ISA/210 (patent family annex) (July 1992)